

Ultrafast laser-induced changes of the magnetic anisotropy in iron garnet films

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Ultrafast laser-induced control of the magnetic state of media is the one of the most intriguing and controversial fields of the contemporal condensed matter physics. Changing magnetic anisotropy by a femtosecond laser pulse is one of the most versatile approaches for controlling the magnetic state on the subnanosecond time scale. In this paper we report on the results of the magneto-optical pump-probe studies of laser-induced magnetization dynamics in low symmetry ferrimagnetic dielectric $(\text{YBiPrLu})_3(\text{FeGa})_5\text{O}_{12}$ garnet films grown on the (210)-oriented $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ substrate. We demonstrate that the femtosecond laser pulse triggers both coherent and incoherent spin dynamics at distinct time scales. Demagnetization mediated by slow magnon-phonon interactions occurs on the time scale of ~ 500 ps, while on a shorter time scale the magnetization precession is excited. Circularly polarized laser pulses excite the magnetization precession via the ultrafast inverse Faraday effect. Furthermore, as we demonstrate experimentally and phenomenologically, the changes of the anisotropy parameters mediated by the lattice heating are triggered by laser pulses of any polarization on a few picosecond time scale and compete with the inverse Faraday effect. We show that the orientation of the external magnetic field with respect to magnetization easy plane noticeably affects the precession excited via the anisotropy change. Importantly, the relative contributions from the inverse Faraday effect and the change of the growth-induced anisotropy can be controlled by varying the applied magnetic field. As a result, the amplitude and the initial phase of the excited magnetization precession can be gradually tuned.

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I. INTRODUCTION

Ferrimagnetic rare-earth iron garnets $R_3\text{Fe}_5\text{O}_{12}$ and related compounds, where R stands for yttrium, rare-earth and some other ions, e.g. bismuth, are highly resistive dielectrics with the bandgap of $E_g \sim 2.8$ eV. These materials have passed through several periods of the strong research interest, triggered by their unique physical properties and important applications.^{1,2} First of all, yttrium iron garnet (YIG) possesses a record-narrow width of the ferromagnetic resonance line³ and the strong magneto-acoustic coupling,⁴ thus being a perfect medium for a large family of microwave devices. Being centrosymmetric cubic in the bulk, epitaxial magnetic garnet films reduce their crystallographic symmetry and generally lose the center of inversion, which allows for various effects forbidden in cubic bulk samples. Good example of such an effect is the optical second harmonic generation.^{5,6} Symmetry breaking enables intrinsic giant linear magneto-electric effect.⁷⁻⁹ Owing to the piezomagnetic response, thin magnetic garnet films are among the building blocs for composite multiferroics as well.¹⁰ High values of Faraday rotation in Bi-substituted YIG allows designing efficient magneto-optical isolators and waveguides.¹¹ The same property makes magnetic garnets the key material for engineering magneto-photonic¹² and magneto-plasmonic¹³ structures. Thin magnetic garnet films with uniaxial anisotropy were among the model media for developing magnetic bubble domain technology.^{14,15} Nowadays, thin YIG films are the model functional media for testing var-

ious concepts of magnonics,¹⁶ owing to their exceptionally low spin waves damping,¹⁷ and novel effects at the dielectric garnet/metal interfaces.¹⁸

This outstanding functionality of the magnetic garnet films originates from the fact, that their magnetization, magnetic anisotropy, compensation points and other properties can be tailored in a wide range. Thus, growth conditions, a type of substrate and a chemical composition allow for fabricating the garnets of easy-plane, out-of-plane, and more intricate types of the anisotropy.^{1,2} Furthermore, the magnetic anisotropy of garnets is highly susceptible to various external stimuli, such as temperature,¹⁹ strain,¹⁰ and optical irradiation.^{20,21} As a result, efficient dynamical modulation of parameters of ferromagnetic resonance and spin waves spectra, domain patterns, etc. can be realized by using these external stimuli. Recently, the control of the magnetic anisotropy of garnet films was demonstrated by means of femtosecond laser pulses, thus showing the feasibility of ultrafast photomagnetic effects.²²

By now, femtosecond laser-induced changes of the effective magnetic anisotropy field has been shown to be one of the most common effects, observed in magnetically-ordered dielectrics,²²⁻²⁴ semiconductors^{26,27} and metals.^{28,29} In all substances ultrafast change of the intrinsic magnetocrystalline and shape anisotropies by femtosecond laser pulses or appearance of a transient anisotropy axis manifest themselves via coherent spin precession, despite very different microscopic mechanisms underlying these processes,³⁰ related, first of all, to the corresponding electronic band structures. For example, excitation

by a femtosecond laser pulse of magnetically ordered metals results in subpicosecond increase of the electronic temperature which determines subsequent dynamics of other subsystem.^{31–33} Therefore, in such media changes of the shape and magnetocrystalline anisotropies mostly result from the ultrafast heating.^{28,29} Magnetically ordered dielectrics, in contrast, under the influence of femtosecond laser pulses demonstrate a variety of both thermal and nonthermal mechanisms leading to the anisotropy changes.

The most prominent nonthermal changes of the magnetic anisotropy in dielectrics under the action of laser pulses were observed in several substituted iron garnets,^{22,34,35} where linearly polarized pulses induce a transient anisotropy axis due to charge-transfer optical transitions. These results demonstrated feasibility to control the magnetic anisotropy by changing the azimuthal angle of the laser pulse polarization. Furthermore, this mechanism allowed achieving relatively high amplitudes of laser-induced magnetization precession.³⁴ Very recently, the laser-induced uniaxial anisotropy mediated by the acoustic phonons has been shown to enhance magnetization of the Cu-based organic-inorganic Heisenberg magnets.²⁴ Rapid heating related to the laser pulses can also lead to a modification of the intrinsic magnetocrystalline anisotropy of dielectrics. Exploring this mechanism in the rare-earth orthoferrites in the vicinity of spin reorientation phase transitions²³ yielded a number of remarkable results on coherent control of magnetization.^{36,37} Evidently, laser-induced thermal changes on the intrinsic magnetic anisotropy should be a general phenomenon in dielectrics, not restricted exclusively to the vicinity of phase transitions. Such a process, therefore, can be an alternative way to the spin waves excitation. Thermal modulation of the anisotropy can affect the spin waves spectrum and other dynamical properties and, therefore, understanding timescales and strength of this effects in dielectrics is important for implementation of laser pulses as excitation stimuli in future magnonic,³⁹ magneto-plasmonic,⁴⁰ spintronic and spin-optronic devices. However, to the best of our knowledge ultrafast thermal effects on anisotropy of dielectrics, except for orthoferrites,^{23,36–38,41} have not been explored so far.

In this paper we report on the results of the experimental studies of ultrafast magnetization dynamics in substituted iron garnet films characterized by a pronounced growth-induced anisotropy of the easy-axis type. We demonstrate that the impact of a femtosecond laser pulse on the magnetic films induces polarization-independent changes of various growth-induced anisotropy parameters. Their relative contributions can be distinguished by analysing the azimuthal field dependencies of the initial phase of the induced precession. We argue that the most plausible mechanism underlying the observed change of the magnetic anisotropy is the lattice heating, which is expected to take place on a picosecond timescale. We show that the amplitude of the magnetization precession

induced via this mechanism is comparable to that induced by the ultrafast inverse Faraday effect. We show that the relative contribution of these two mechanisms to the excitation of the magnetization precession can be tuned by changing the value of the applied magnetic field. Importantly, this allowed us to vary gradually the initial phase of the magnetization precession in the range of $\sim \pi/2$, which is defined by the excitation mechanism. Along with the anisotropy change and the ultrafast inverse Faraday effect, laser pulses trigger the demagnetization, which characteristic time is on the order of ~ 500 ps. Due to this relatively large time the laser-induced demagnetization in garnets does not contribute to the excitation of the precession.

This paper is organized as follows. In Sec. II we introduce phenomenological description of the magnetic anisotropy of the substituted iron garnet films grown on (210) substrate. We consider how the ultrafast change of the magnetic anisotropy parameters is expected to manifest itself in the experiment. Sec. III is dedicated to the magneto-optical characterization of the magnetic garnet films chosen for the study, and in Sec. IV we present the details of the magneto-optical pump-probe experiments. In Sec. V A we present experimental results on magnetization dynamics in the (210) garnet film with strong out-of-plane anisotropy. Based on these results we deduce the parameters of the laser-induced demagnetization. In Sec. V B the experimental data on the magnetization dynamics after the laser pulse excitation in the (210) garnet film with moderate anisotropy is discussed. This is followed by the analysis of two mechanisms of the precession excitation, the ultrafast inverse Faraday effect (Sec. V C) and change of the growth-induced anisotropy parameters under the influence of laser pulses (Sec. V D).

II. MAGNETIC ANISOTROPY OF A GARNET FILM GROWN ON (210)-ORIENTED SUBSTRATE

Substituted iron garnet films are characterized by the magnetic anisotropy which is the consequence of the interplay between cubic anisotropy inherent to their crystallographic structure and the growth-induced one. The latter is dependent on several factors such as substrate lattice parameters and crystallographic orientation, particular chemical composition and parameters of the growth technology (for a review see e.g.[14]). Typically in films grown on low symmetry substrates the growth-induced contribution strongly dominates resulting in the easy-axis anisotropy.

We consider here a magnetic garnet film grown on the (210)-substrate. The reference frame is chosen as shown in Fig.1(a). The x axis is directed along the [001] crystallographic direction. From the symmetry point of view this direction is the $\bar{2}_x$ axis and the crystallographic point group of this film is m .⁶ The z -axis is directed along the [210] crystallographic axis and is normal to the sample

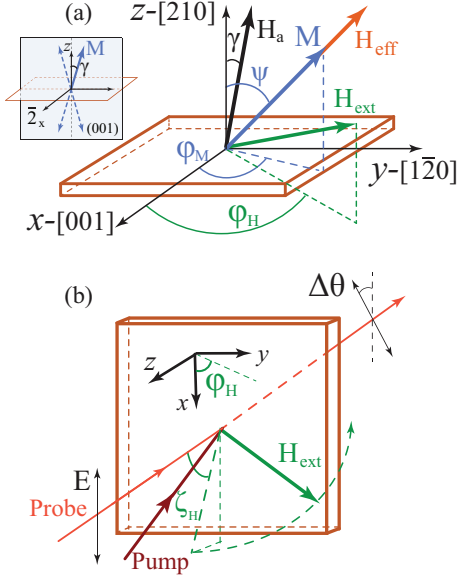


FIG. 1: (Color online) (a) Orientations of the crystallographic axes of the (210)-film, the applied magnetic field \mathbf{H}_{ext} , the effective anisotropy field \mathbf{H}_a , and the net effective field \mathbf{H}_{eff} . x, y, z axes are directed along [001], [120], and [210] crystallographic axes, respectively. Effective anisotropy field \mathbf{H}_a makes an angle $\gamma = 16^\circ$ with the z -axis in the (yz) -plane, as shown in the inset.⁷ The magnetic field direction is described by the polar angle $\zeta_H = 80^\circ$ and the azimuthal angle φ_H . The direction of magnetization \mathbf{M} is described by the polar ψ and azimuthal φ_M angles. In a general case the magnetization \mathbf{M} is not collinear with the applied magnetic field and is not in the (yz) -plane. (b) Geometry of the pump-probe experiment (see text for the details).

plane. The magnetic anisotropy energy of such a film can be expressed as^{14,42}

$$w_a = K_u m_z^2 + K_i m_y^2 + K_{yz} m_y m_z + w_{\text{cub}}, \quad (1)$$

where $m_k = M_k/M_S$ ($k = x, y, z$) are the normalized components of the magnetization, M_S is the saturation magnetization. In this expression w_{cub} is the cubic anisotropy energy, K_u and K_i are the uniaxial out-of-plane and in-plane parameters respectively, K_{yz} is the orthorhombic anisotropy coefficient. Since the cubic anisotropy w_{cub} is typically much weaker than the

growth-induced one, the former can be neglected in the following consideration. Clearly, the equilibrium orientation of the magnetization is determined by the ratios between growth-induced anisotropy parameters entering Eq.(1). In particular, the yz plane is the easy plane of the magnetization. The orientation of the magnetization in this plane is determined by the values of K_u , K_i and K_{yz} for a particular film.

We can write the expression for the effective growth-induced anisotropy field \mathbf{H}_a in a form

$$\mathbf{H}_a = -\frac{\partial w_a}{\partial \mathbf{M}} = \frac{1}{M_S^2} \begin{pmatrix} 0 \\ (-2K_i M_y - K_{yz} M_z) \\ (-2K_u M_z - K_{yz} M_y) \end{pmatrix}. \quad (2)$$

From this expression one can see that a laser-induced change of the anisotropy parameters K_u , K_i or K_{yz} should lead to changes of both the value and the direction of the effective anisotropy field. In order to illustrate how such changes of the magnetic anisotropy coefficients can lead to the excitation of the magnetization precession we employ the phenomenological approach based on the Landau-Lifshitz equation⁴³

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} = \gamma \mathbf{M} \times (\mathbf{H}_{\text{ext}} + \mathbf{H}_a + \mathbf{H}_d), \quad (3)$$

where γ is the gyromagnetic ratio, \mathbf{M} is the magnetization, and \mathbf{H}_{eff} is the net effective magnetic field, which consists of the applied magnetic field \mathbf{H}_{ext} , the internal anisotropy field \mathbf{H}_a , and the demagnetizing field $\mathbf{H}_d = -4\pi M_S \mathbf{z}$. In Eq. (3) the term responsible for the precession damping is omitted, since we focus on a time scale when its contribution can be ignored.

In this approach the effect of the femtosecond laser pulse is introduced into the expression for the effective magnetic field \mathbf{H}_{eff} by adding new terms and modifying existing ones. In particular, (i) the change of the anisotropy constants $\Delta K_u(t)$, $\Delta K_i(t)$, and $\Delta K_{yz}(t)$ manifests itself in the change of the effective anisotropy field $\Delta \mathbf{H}_a(t)$; (ii) the effective field $\mathbf{H}_{\text{om}}(t)$ accounts for the opto-magnetic effects, such as the ultrafast inverse Faraday and Cotton-Mouton effects.^{44–46}

Using Eqs.(2, 3) one can derive general expressions for the torque acting on the magnetization due to these effects:

$$\frac{1}{\gamma} \frac{d\mathbf{M}}{dt} = \mathbf{M} \times (\Delta \mathbf{H}_a + \mathbf{H}_{\text{om}}) = \begin{pmatrix} (2(\Delta K_i - \Delta K_u) m_y m_z + \Delta K_{yz}(m_z^2 - m_y^2)) \\ (2\Delta K_u m_x m_z + \Delta K_{yz} m_x m_y) \\ (-2\Delta K_i m_x m_y - \Delta K_{yz} m_x m_z) \end{pmatrix} + \begin{pmatrix} (H_{\text{om}z} M_y - H_{\text{omy}} M_z) \\ (H_{\text{om}x} M_z - H_{\text{om}z} M_x) \\ (H_{\text{omy}} M_x - H_{\text{omx}} M_y) \end{pmatrix} \quad (4)$$

Importantly, Eq.(4) clearly demonstrates that the strength and the direction of the torque acting on

the magnetization due to the induced changes of any anisotropy constants ΔK as well as due to ultrafast opto-

magnetic effects are determined by the initial orientation of the magnetization.

Dependence of the torque (4) on the initial magnetization direction makes iron garnet films grown on (210)-oriented substrates the model media to explore experimentally laser-induced changes of the magnetic anisotropy parameters ΔK . Once the out-of plane orientation of the magnetization is defined by the magnetic anisotropy, the external magnetic field \mathbf{H}_{ext} of tunable strength applied along the hard axis provides gradual change of the equilibrium orientation of the magnetization. Therefore, this should favor experimental detection of the magnetization dynamics excited due to the changes of the anisotropy parameters and, moreover, can allow one to determine their relative contributions. It is also important to emphasize that the character of the magnetization dynamics excited due to the such changes would be strongly dependent on the time scale of the involved processes. Generally speaking, it should occur on a time scale shorter than the characteristic time of the magnetization precession. If it is not the case, then one can expect slow deflection movement of the magnetization towards a new equilibrium position instead of the precessional motion.

We note that, when deriving the expression for the torque (4), we neglected a possible change of the saturation magnetization $\Delta M_S(t)$ due to the laser-induced demagnetization.^{47,48} This is justified because the demagnetization in dielectrics occurs on a time scale of several hundreds of picoseconds, which is longer than the typical period 100 ps of precession in magnetic garnet films. Therefore, the demagnetization, although resulting in the changes of demagnetizing field $\Delta \mathbf{H}_d$, is not expected to contribute to the torque (4). The validity of this assumption was verified experimentally as explained in Sec. V.

III. SAMPLES

For investigating the interaction of femtosecond laser pulses with magnetic garnet films manifested in the modification of their parameters we have chosen substituted iron garnet film $(\text{YBiPrLu})_3(\text{FeGa})_5\text{O}_{12}$ grown by liquid phase epitaxy method on (210)-oriented substrates of gadolinium gallium garnet $\text{Gd}_3\text{Ga}_5\text{O}_{12}(\text{GGG})$. The films thickness was of 10 μm . X-ray diffraction characterization yielded the lattice constants of 12.5322 Å and 12.4844 Å for the garnet film and the GGG substrate, respectively. The lattice mismatch between the film and the substrate, therefore, was of +2.65%. Effective anisotropy easy axis is oblique in the yz plane with the angle of 16° to the sample normal,⁷ as shown in Fig. 1(a). This film is characterized by the moderate magnetic anisotropy and will be referred to as the MA-film. The second sample was a film $(\text{YBiPrLu})_3(\text{FeGa})_5\text{O}_{12}$ of similar chemical composition, grown on the (210)-substrate but characterized by a strong growth-induced anisotropy

(SA-film).

Magnetic, optical and magneto-optical properties of the films were characterized by means of magneto-optical Faraday rotation measurements at wavelength of 690 nm. At this wavelength samples possess optical absorption coefficient of $\alpha \simeq 400 \text{ cm}^{-1}$. This value is, on the one hand, much lower than absorption in magnetic metals where thermal effects dominate in ultrafast dynamics, but, on the other hand, is definitely not negligible. Therefore, thermal effects occurring as a result of femtosecond laser pulse impact should be taken into account. The rotation angle θ_F of the polarization plane of the light propagating along the sample normal was measured as a function of the DC magnetic field H_{ext} . The field was applied at 80° to the sample normal (10° with respect to the sample plane, see inset in Fig. 2(a)). As one can see from Fig. 2(a) in the fields up to 0.63 T the magnetization of the SA-film was not noticeably inclined from the easy anisotropy axis, confirming the strong growth-induced anisotropy. On the contrary, in the MA-film the angle between the magnetization and the easy axis was gradually increasing up to 45° in the applied field from 0 to 0.63 T (see Fig. 2(b)). In order to estimate the magnetic anisotropy strength of the studied samples we performed ferromagnetic resonance measurements. However, due to the large FMR linewidth no reliable numbers could be extracted from the measurements. The Faraday rotation θ_s at remanence is of 3.8° and 9.3° in the MA- and SA-films, respectively (see Fig. 2).

IV. EXPERIMENTAL

All-optical pump-probe experiments were performed employing technique analogous to that described elsewhere.³⁰ Optical parametric amplifier pumped by the femtosecond regenerative $\text{Yb} : \text{KGd}(\text{WO}_4)_2$ amplifier produced 170 fs laser pulses with repetition rate of 5 kHz, and the central wavelength of $\lambda=690 \text{ nm}$, which we used as both pump and probe. The measurements were done in transmission geometry, as shown in Fig. 1(b). The pump-induced magnetization dynamics was monitored by measuring the Faraday rotation for the probe pulses as a function of the pump-probe time delay t . Rotation of the probe polarization plane $\Delta\theta$ is proportional to the change of the component normal to the film plane. Therefore, $\Delta\theta$ normalized with respect to the static Faraday rotation θ_s at remanence (Fig. 2) is the measure of the laser-induced deviation of the magnetization from its equilibrium orientation. Pump pulses were either linearly or circularly polarized. The angle of incidence for the pump pulses was of 12°. Pump fluency was of 2 mJ/cm². Linearly polarized probe pulses propagates along the z -axis. Probe fluency was ~ 50 times lower than that of the pump. The external magnetic field \mathbf{H}_{ext} of up to 0.63 T was applied at 80° to the sample normal in order to deflect the magnetization from its easy axis. All measurement were performed at $T=295 \text{ K}$.

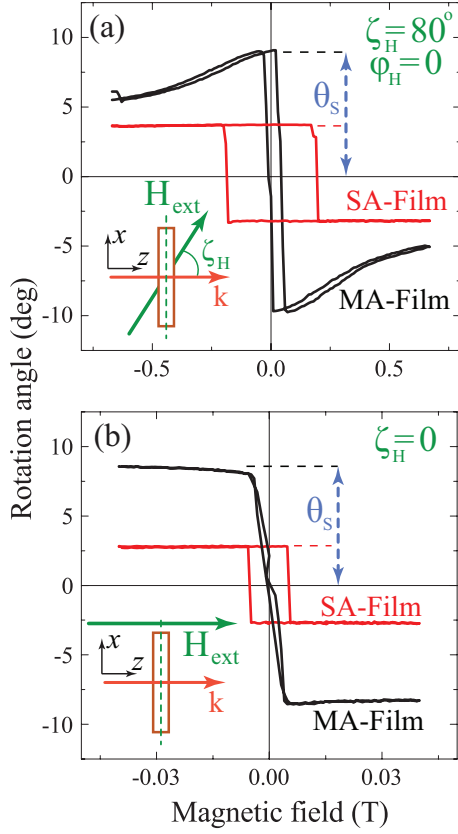


FIG. 2: (Color online) Static Faraday rotation as a function of the external magnetic field in the SA-film (red line) and MA-film (black line) with external field applied at (a) $\zeta_H = 80^\circ$ and (b) $\zeta_H = 0$. θ_s denotes the Faraday rotation at remanence, which is proportional to the samples magnetization M_s . Insets show the experimental geometries used for measuring the static Faraday rotation in the samples.

In our experiments the dynamics of the magnetization following the femtosecond laser pulse excitation was studied as a function of the sign and magnitude of the applied magnetic field $\pm H_{\text{ext}}$ and of the pump polarization. In order to distinguish effects arising due to the helicity σ^\pm $\Delta\theta_{\text{hd}}$ and those dependent on the H_{ext} sign $\Delta\theta_{\text{fd}}$ we will use the expressions

$$\frac{\Delta\theta_{\text{hd}}}{\theta_s} = \frac{\Delta\theta(\sigma^+; +H) - \Delta\theta(\sigma^-; +H)}{2\theta_s}; \quad (5)$$

$$\frac{\Delta\theta_{\text{fd}}}{\theta_s} = \frac{\Delta\theta(\sigma^+; +H) - \Delta\theta(\sigma^+; -H)}{2\theta_s}. \quad (6)$$

All the pump-probe data were normalized to the static Faraday rotation θ_s at remanence (Fig. 2(b)) allowing one to evaluate the relative change of the \mathbf{M}_z component.

V. RESULTS AND DISCUSSION

A. Laser-induced demagnetization

In laser-induced dynamics two kinds of effects are mostly observed. The first is the demagnetization and the second is the excitation of the precession of magnetization. The demagnetization itself occurring on the relatively short timescale, for instance in metals, can be a driving mechanism of the excitation of the magnetization precession. Firstly we investigate the magnetization change under the action of femtosecond laser pulses often referred to as a laser-induced demagnetization in order to specify the strength and the time scale of this process in the garnet films under study. For this we selected the SA-film characterized by the strong easy axis anisotropy where the magnetization is directed along the sample normal. Available external field applied at the 80° with respect to the sample normal is not sufficient to deflect the magnetization from the easy axis (Fig. 2(a)). As can be seen in Fig. 1(b), in such case we probe solely the change of the magnetization magnitude while other processes such as magnetization precession are not detected even if excited. Therefore, such experimental geometry is the most suitable for detecting laser-induced demagnetization via the Faraday rotation.

Dependencies of the polarization rotation of the probe pulse as a function of the pump-probe time delay t are shown in Fig. 3(a) for the two opposite applied magnetic fields exceeding the saturation field. Fig. 3(a) shows that the dynamics notably changes as the \mathbf{H}_{ext} is reversed. In order to analyse this behavior we extracted the sign-dependent contribution (6) from the measured signals, which can be reliably considered as a measure of the pump-induced change of M_z . We note that such approach excludes the sign-independent contribution. Its time evolution closely resembles the temporal transmission changes $\Delta T/T$ caused by pump shown in Fig. 3(c). Therefore, we attribute this $\Delta\theta(t; \pm H)$ contribution to the changes of optical or magneto-optical sample properties but not to the alteration of the magnetization magnitude or its orientation.

The time-delay dependence of the signal $\Delta\theta_{\text{fd}}(t)/\theta_s \sim \Delta M_z(t)/M_s$ has been fitted by a function

$$\frac{\Delta\theta_{\text{fd}}(t)}{\theta_s} = \frac{\theta_{\text{dm}}}{\theta_s} e^{-t/\tau_{\text{dm}}} - 1, \quad (7)$$

yielding the characteristic time $\tau_{\text{dm}} = 500 \pm 5$ ps. The value $\theta_{\text{dm}}/\theta_s$ characterizes the magnitude of this process and amounts to 0.2%. Noting that the magnetization is directed along the sample normal, we conclude from the experimental data that the action of the pump pulses induces slow exponential change of the magnetization magnitude.

The long time scale of 500 ps and small amount of 0.2% of the observed change of the magnetization suggest that pump pulses trigger the demagnetization in

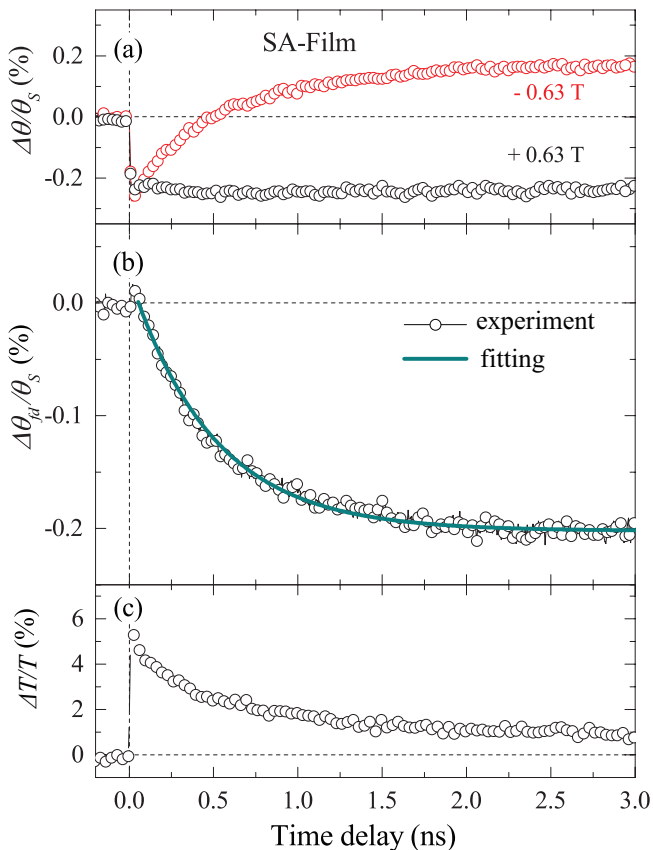


FIG. 3: (Color online) (a) Normalized rotation of the probe polarization induced by the linearly polarized laser pulse as a function of the pump-probe time delay t measured for positive and negative magnetic field of ± 0.63 T in the SA-film. (b) Field-dependent contribution to the laser-induced dynamics, extracted using Eq. (6) from the data shown in the panel (a). Solid line is a fit using the Eq.(7). (c) Normalized change of the sample transmission as a function of pump-probe time delay t .

the sample. Such a process in magnetically-ordered dielectrics was reported in Ref. 47 and studied for the case of ferro- and ferrimagnetic,^{48,50} weak ferromagnetic,⁴⁷ and antiferromagnetic⁴⁹ materials. This process can be understood as follows. Optical absorption of a laser pulse leads to excitation of electrons to the 3d sublevels of the Fe^{3+} ions which decays at the femtosecond timescale leading to nonequilibrium phonon excitation and subsequent increase of the lattice temperature.⁵⁷ Relatively weak phonon-magnon interaction mediates energy transfer from lattice to incoherent magnons increasing the spin temperature appearing as a decrease of magnetization magnitude.

We would like to note, that the slow change of the magnetization value can, in general, lead to the deviation of the magnetization from its equilibrium orientation resulting in a change of M_z . Indeed, the orientation of the magnetization is defined by the balance between the magnetic anisotropy energy w_a , the shape anisotropy en-

ergy $-4\pi M_z^2$, and the Zeeman energy $-\mathbf{M} \cdot \mathbf{H}_{\text{ext}}$. While the anisotropy energy depends on the normalized components m_k of the magnetization but not on their absolute values (see Eq. (1)), the two other energies would decrease as the absolute magnitude of the magnetization decreases. As a result, the relative contribution of the magnetic anisotropy w_a grows. If the easy-axis anisotropy energy is comparable to the Zeeman energy the demagnetization would lead to the increase of M_z competing with decrease of M_S . The described process is not expected to give any noticeable contribution in the film under consideration due to the dominance of the easy-axis anisotropy.

B. Laser-induced magnetization precession

Having determined the characteristic time and strength of the induced demagnetization in the SA-film, we now discuss the magnetization dynamics caused by the pump in the garnet film with the moderate growth-induced anisotropy (MA-film). Fig. 4(a) shows the rotation of the probe polarization induced by the circularly polarized laser pulse as a function of the pump-probe time delay for different values of the magnetic field H_{ext} . Clear oscillations of the probe polarization are observed superimposed on a slowly changing background. The frequency f of the oscillations increases with the field (Fig. 4(b)). This indicates that the observed oscillations result from the precession of the magnetization triggered by the pump pulses. Fig. 4(c) shows precession frequency f as a function of the azimuthal angle φ_H of the external field $H_{\text{ext}}=0.26$ T (see Fig. 1(a)). From these results we were able to determine the orientation of the hard magnetization x axis of the sample. In the following discussion we adopt the frame of reference, where $\varphi_H=0$ corresponds to the projection of the H_{ext} on the sample plane being parallel to the x axis (Fig. 1(b)).

The central goal of our study is to explore the possibility of excitation of the magnetization precession via laser-induced change of the anisotropy parameters (see Eq.(4)). To this end most of the following studies were performed for the field H_{ext} directed close to the sample's hard axis ($\xi = 80^\circ$, $\varphi_H = 0$). In this geometry, when the field makes a small angle of 10° with the hard magnetization axis (x axis), a change of any of the anisotropy constants ΔK is expected to affect the orientation and the value of the effective field $\mathbf{H}_{\text{eff}} = \mathbf{H}_a + \mathbf{H}_d + \mathbf{H}_{\text{ext}}$, which in turn should result in the finite torque \mathbf{T} according to Eq.(4).

For revealing possible mechanisms of the laser-induced precession and the slowly varying background signal we have studied the influence of the pump pulses polarization and the sign of H_{ext} on parameters of the probe dynamics. In Fig. 5(a) we plot the time-resolved dynamics of the probe polarization rotation induced by the right- (σ^+) and left-handed (σ^-) pump pulses. Fig. 5(b) shows the dynamics induced by the σ^- -polarized pump pulses

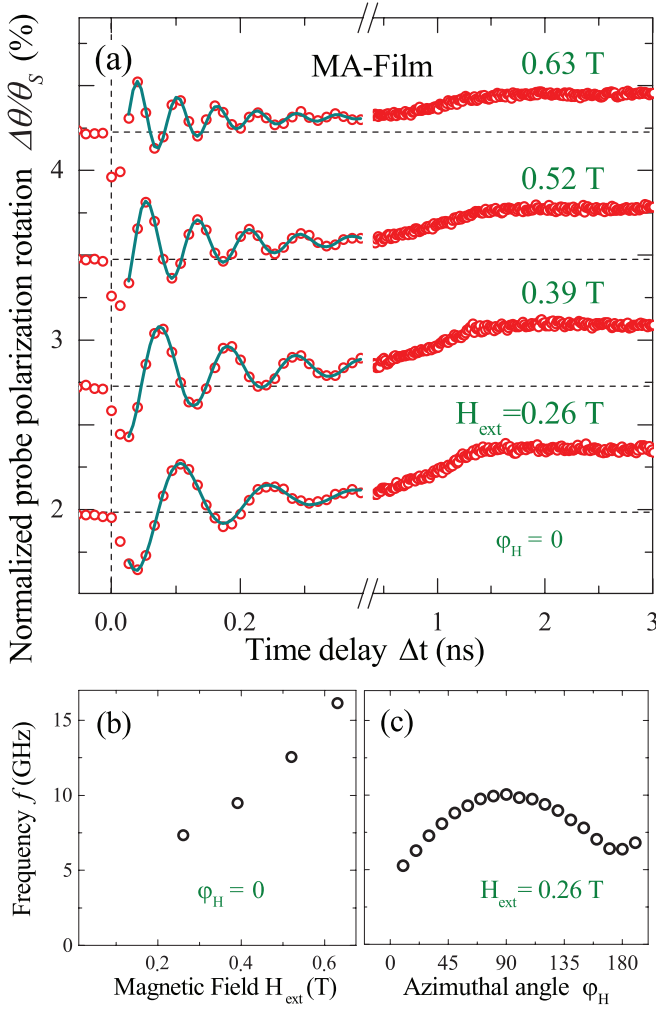


FIG. 4: (a) Normalized rotation of the probe polarization induced by the left-handed (σ^-) circularly polarized pump pulses in the MA-film, measured as a function of the time delay t for various magnitudes of the applied magnetic field. (b) Frequency of oscillations shown in the panel (a) as a function of the magnetic field magnitude (azimuthal angle $\phi_H=0$). (c) Frequency of oscillations shown in the panel (a) as a function of the azimuthal angle ϕ_H .

Such a complex behavior clearly indicates that there are two competing mechanisms responsible for the precession excitation. The first one is sensitive to the helicity of the exciting pulse but not to the applied field sign. The second mechanism is independent from the helicity of the laser pulse but is sensitive to the sign of the applied field. In the following discussion we refer to these two mechanisms as to the helicity-dependent and field-dependent ones. The relative contributions of these two mechanisms depend on the applied magnetic field H_{ext} . This is demonstrated in Fig. 6(b) where we plot the amplitudes $\Delta\theta_{\text{pr}}/\theta_s$ of the helicity dependent and field dependent contributions to the probe polarization

measured in the positive and negative applied fields of various strength. One can see that the change of the pump pulse helicity clearly affects the initial phase of the oscillations in a nontrivial way. In order to evaluate the change of the precession parameters we have fitted the experimental curves shown in Fig. 5(a) by a function

$$\frac{\Delta\theta(t)}{\theta_s} = \frac{\Delta\theta_{\text{pr}}}{\theta_s} e^{-t/\tau_{\text{pr}}} \cos(2\pi ft + \xi_0) + P_2(t), \quad (8)$$

where $\Delta\theta_{\text{pr}}/\theta_s$ is the normalized oscillations amplitude, ξ_0 is the initial phase, τ_{pr} is the oscillations decay time, and $P_2(t)$ is the second-order polynomial function accounting for the slowly varying background to be discussed below. In Fig. 6(a) we plot the initial phase of the precession as a function of the H_{ext} . As one can see, the initial phase ξ_0 takes intermediate values, and, furthermore, depends on both the sign and strength of the applied field and the pump polarization. Thus, in the limit of large fields the switching of the pump pulse helicity leads to the π -change of the initial phase of oscillations. As the applied field decreases this change of the initial phase decreases and almost vanishes in the field of 0.26 T. In contrast, when the pumping takes place with the same polarization but the H_{ext} is reversed then the phase shift vanishes at high fields ± 0.63 T and arises almost to the π -change at low fields.

oscillations (Eqs. (5, 6)).

C. Ultrafast inverse Faraday effect

We showed above that the helicity-dependent contribution to the laser-induced magnetization precession becomes larger as the applied field grows. This indicates that the contribution is stronger when the equilibrium magnetization orientation becomes closer to the sample plane, i.e. almost perpendicular to the pump wave vector (see Fig. 1(b)). At the high-field limit ($H_{\text{ext}} = 0.63$ T) the initial phase of the oscillations of the

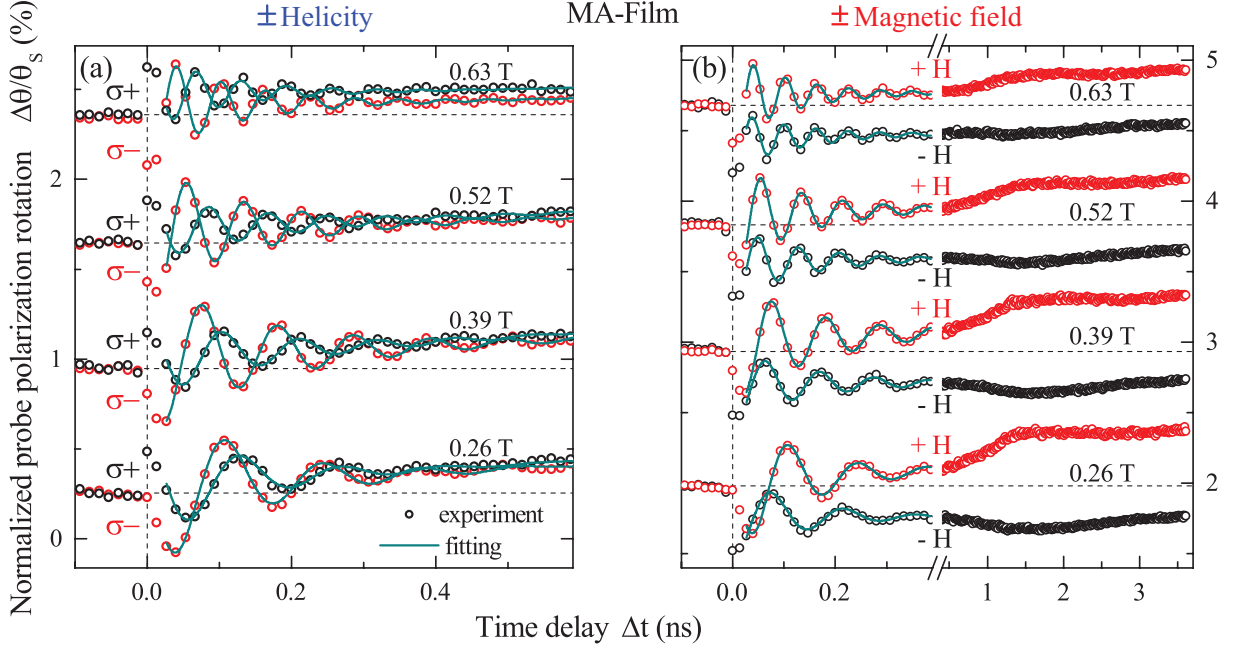


FIG. 5: (a) Normalized probe polarization rotation induced by the left- (σ^-) and right-handed (σ^+) circularly polarized pump pulses in the MA-film measured as a function of the time delay t for the various magnetic fields. (b) Probe polarization rotation induced by the σ^+ -polarized pump pulses as a function of the time delay t measured for the various strength of positive and negative magnetic field $\pm H$.

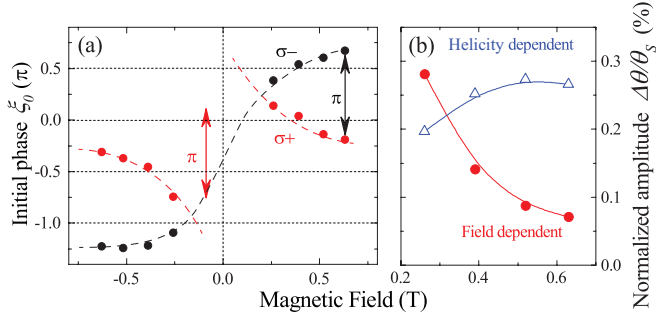


FIG. 6: (a) Initial phase ξ_0 of the probe polarization oscillations excited by σ^\pm -pump pulses in the MA-film as a function of the applied field, as extracted from the experimental data in Fig. 5. (b) Amplitude of the polarization- and field-dependent contribution to the probe polarization oscillations in the MA-film shown in Fig. 5 as a function of the applied magnetic field strength.

probe polarization being proportional to M_z is changed by a π angle when the helicity of the exciting pulse is reversed. This allows us to draw the conclusion, that the observed excitation of the magnetization precession occurs via the ultrafast inverse Faraday effect (IFE).⁴⁴ This effect microscopically originates from the impulsive stimulated Raman scattering on magnons,^{46,51} and can be described as a femtosecond pulse of an effective magnetic field induced by the circularly polarized laser pulse^{44,52,53}

$$\mathbf{H}_{\text{IFE}} \sim \alpha \mathbf{E}(\omega) \times \mathbf{E}(\omega)^* \sim p \alpha I_0 \mathbf{z}, \quad (9)$$

where $\mathbf{E}(\omega)$ is the electric field of the light, α is the magneto-optical susceptibility which also defines the Faraday rotation and I_0 is the pump intensity. The degree of ellipticity p spans from -1 to +1 when the polarization of the pump pulse is tuned from left- to right-handed circularly polarized. \mathbf{H}_{IFE} is directed along the wavevector of the pump pulse close to the z -axis in our experimental geometry (Fig. 1(b)).

In Table I we show the components of the torque created via IFE, derived using Eqs. (4,9), as functions of the polar ψ and azimuthal φ_M angles of the magnetization. As one can see, the torque increases as the angle ϕ between the equilibrium orientation of magnetization and the z -axis increases. In other words, the stronger are $M_{x,y}$ components the stronger is the created torque

(4). This agrees well with the experimental data. Furthermore, it can be shown^{44,46} that the reversal of the applied magnetic field does not affect the initial phase of M_z oscillations excited via IFE. This is also in accordance with our experimental data in the high field limit (see Fig. 5(a) and Fig. 6(a)).

TABLE I: Components of the torque \mathbf{T} arising due to the ultrafast inverse Faraday effect \mathbf{H}_{IFE} and the laser-induced changes of the anisotropy parameters ΔK , as functions of the polar and azimuthal angles ψ and φ_M , describing the equilibrium orientation of the magnetization.

Driving mechanism	T_x	T_y	T_z
\mathbf{H}_{IFE}	$\sin \psi \sin \varphi_M$	$\sin \psi \cos \varphi_M$	0
ΔK_u	$-\sin 2\psi \sin \varphi_M$	$\sin 2\psi \cos \varphi_M$	0
ΔK_i	$\sin 2\psi \sin \varphi_M$	0	$-\sin^2 \psi \sin 2\varphi_M$
ΔK_{yz}	$\cos^2 \psi - \sin^2 \psi \sin^2 \varphi_M$	$\frac{1}{2} \sin^2 \psi \sin 2\varphi_M$	$-\frac{1}{2} \sin 2\psi \cos \varphi_M$

As follows from the expression given in Table I, the torque acting on the magnetization due to the IFE field has only x and y components. Its direction defines the initial phase of the magnetization precession. In order to compare calculation with the experimental data, we recall here that the oscillations of the probe polarization are the fingerprint of the M_z state. Furthermore, we would like to note that interpretation of the initial phase ξ_0 of cosine-like function (8) depends on the mechanism of the precession excitation.⁴⁶ When the spin system is affected only during the pump pulse action the value of the initial phase $\xi_0 = \pi/2$ in Eq. (8) means that the magnetization was deflected by the laser-induced torque directed perpendicular to the z -axis.

D. Laser-induced change of the magnetic anisotropy

As opposed to the inverse Faraday effect discussed in the previous section the effectiveness of the second mechanism goes down as the applied field grows. Circular polarization is not a prerequisite for such excitation. Previous studies have shown that the linearly polarized femtosecond laser pulse can induce transient anisotropy axis in iron garnets and thus excite the magnetization precession.^{22,25,34,35,50,54} In order to elucidate possible microscopic mechanism of the laser-induced precession sensitive to the sign of the applied field, we have verified whether the precession can be excited by the linearly polarized laser pulses. Furthermore, we have checked whether the azimuthal angle of the pump polarization affects the excitation process. As shown in Fig. 7(a), the precession can be effectively excited by the linearly polarized pulses. Nevertheless, the initial phase as well as the amplitude of the excited precession are essentially independent from the pump polarization angle. Therefore, the magnetization precession in the film under study is excited by both circularly and linearly polarized pump pulses. Moreover, in striking contrast to the results reported in [34,35,50,54] the precession changes negligibly as the azimuthal angle ϕ of the linear polarization of the pulses varies. Based on these results we argue that the precession excitation occurs via fast laser-induced

changes of the growth-induced anisotropy parameters, but not via induction of the transient anisotropy axis.

In order to test this hypothesis and to distinguish contributions from different anisotropy parameters ΔK_u , ΔK_i , and ΔK_{yz} , we have studied the laser-induced excitation of the precession in the magnetic field of $H_{\text{ext}} = 0.26$ T applied at different azimuthal angles in the range $-90^\circ < \varphi_H < 90^\circ$.⁵⁵ A low strength of the applied field has been chosen in order to maximize the contribution to the precession excitation originating from the field-dependent mechanism. Fig. 7(b) shows the initial phase ξ_0 of the precession as a function of the azimuthal angle φ_H . As one can see, ξ_0 is gradually changing by more than $\pi/2$ in the range $-80^\circ < \varphi_H < 20^\circ$. Furthermore, there is an abrupt jump of ξ_0 at $\varphi_H = -80^\circ$. This experiment clearly demonstrates that the initial phase of the precession is very sensitive to the orientation of the applied magnetic field, i.e. to the equilibrium orientation of the magnetization with respect to the sample easy axis.

We note that the change of the anisotropy parameters can be considered as a displacing mechanism of the excitation of the precession when the spin system reacts to the changes of H_{eff} direction under the impact of the laser pulse. In this case the phase $\xi_0 = 0$ of the oscillations of the M_z -component of the magnetization indicates that the corresponding initial torque vector is directed in the xy plane, i.e. the z component of \mathbf{H}_{eff} has been altered. The phase $\xi_0 = \pi/2$ corresponds to the situation, when the excitation leads to the change of the component of the effective field in the xy plane. Consequently, the laser-induced torque acting on the magnetization is directed along the z axis.

In the Table I we present the components of the torque (4) as a function of the polar ψ and azimuthal φ_M angles of the magnetization. These expressions allow us to eliminate the change of the K_u parameter as a major driving mechanism for the precession excitation. Indeed, for any orientation of the magnetization the T_z -component related to the ΔK_u vanishes. This means that no pronounced azimuthal dependence of the initial phase ξ_0 of the M_z -oscillations is expected. This strongly contradicts the data in Fig. 7(b), where the initial phase ξ_0 spans over a range exceeding $\pi/2$, thus indicating that at least for some directions of the applied magnetic field

the T_z -component is nonzero.

Another important conclusion which can be drawn from the results shown in Fig. 7(b) is that the initial phase ξ_0 is close to zero when the azimuthal angle is $\varphi_H = 90^\circ$. Note, that in this case both the effective anisotropy field \mathbf{H}_a and the applied magnetic field lie in the yz plane (see Fig. 1(a)), which is the easy plane for the magnetization. In this geometry, the net effective field \mathbf{H}_{eff} should remain in the yz plane even if the laser-induced changes of any of the anisotropy parameters ΔK occur. This, in turn, corresponds to the initial phase $\xi_0 = 0$ of the laser-induced precession, in agreement with that observed in the experiment.

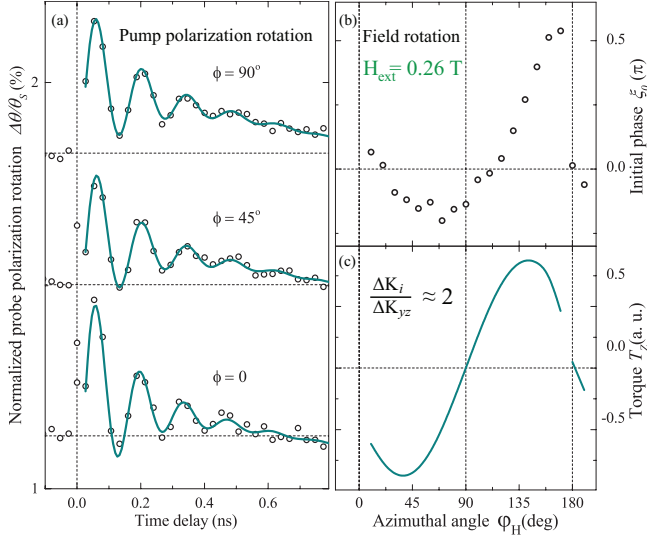


FIG. 7: (a) Normalized probe polarization rotation induced by the linearly polarized pump pulses of different azimuthal angles ϕ measured as a function of the time delay t . (b) Initial phase ξ_0 of the laser-induced precession as a function of φ_H measured for external magnetic field of $H_{\text{ext}} = 0.26$ T in the MA-film. (c) T_z component of the torque (4) calculated as a function of φ_H for a case when ΔK_i is twice as large as ΔK_{yz} . Note, that the abrupt change of the initial phase ξ_0 observed at $\varphi_H = -80^\circ$ occurs because the effective anisotropy field H_a jumps to another equilibrium position when the polar angle $\psi_H < -80^\circ$.

In order to further substantiate suggested mechanism of the precession excitation, we have modeled the changes of the different components of the torque (4) as functions of either K_i or K_{yz} modified under the action of the pump pulse. The calculations were performed taking realistic anisotropy parameters that adequately describe the fact that the equilibrium easy axis of the magnetization lies in the yz plane and makes the angle of 16° with the z axis.⁷ The equilibrium orientation of the magnetization (ψ , φ_M) has been calculated using these parameters for each direction of the H_{ext} in the range of $-90^\circ < \varphi_H < 90^\circ$. Then the laser-induced torque (4) has been calculated assuming changes of K_i or K_{yz} anisotropy parameters. For the sake of clarity the torque occurring due to the ultrafast inverse Faraday effect was

neglected. Fig. 7(c) shows T_z -component of the torque (4) as a function of φ_H for the case when the laser-induced decrease of the orthorhombic anisotropy parameter ΔK_{yz} is twice as large as the change of the in-plane uniaxial parameter ΔK_i . As one can see, the outcome of this model agrees with the experimental results. In fact, the main features of the variations of the T_z -component of the torque (Fig. 7(c)) correlate with the changes of the initial phase of the precession ξ_0 (Fig. 7(b)). In Fig. 8(c) we illustrate the excitation of the precession in two distinct situations for $\varphi_H = 0$ and $\varphi_H = 90^\circ$. We note that although the T_z -component of the torque vanishes for certain angles φ_H the total torque remains always nonzero. This is supported by the experimental observations, that for any azimuthal angle in the range $-90^\circ < \varphi_H < 90^\circ$ the magnetization precession is still excited.

The described above analysis allows us to conclude that the field-dependent excitation of the magnetization precession occurs via the ultrafast change of the growth-induced anisotropy parameters. Since this change is independent from the laser pulse polarization, we argue that the heating can be a plausible mechanism underlying the process. Indeed, various coefficients of the growth-induced anisotropy are known to be temperature dependent.⁵⁶ Thus, the increase of the lattice temperature in magnetic dielectrics on a picosecond time scale following the absorption of the fraction of the femtosecond laser pulse energy, should affect the magnetic anisotropy on the same time scale. The growth-induced anisotropy parameters exhibit distinct temperature dependencies, and, as a result, they change differently in response to the heating. The subsequent cooling of the lattice relies on the much slower heat dissipation. Consequently, the relaxation of the effective anisotropy field to its equilibrium value is expected to be the slow process, in agreement with the displacing character of the change of the anisotropy parameters.

We note that the polarization-independent excitation of the magnetization precession in garnets has been also reported in [25,50]. Both studies were carried out on the films grown on high symmetry (001) GGG substrates and possessing easy-plane-type anisotropy, with no pronounced in-plane anisotropy. In [50] it has been shown that the polarization dependent contribution to the precession excitation occurs with magnetic field being deflected from the sample plane. The authors have interpreted the mechanism of the precession excitation as the nonthermal photo-induced anisotropy. Nevertheless, we suggest that the thermal change of the anisotropy could also contribute to the excitation process. However, distinguishing this mechanism from the others would be challenging, since the (001) films do not allow for experimental studies, where the torque occurring due to the laser-induced thermal changes of magnetic anisotropy possesses strong dependence on the applied field orientation (Fig. 7). In [25] the excitation of the precession was realized in the geometry when the magnetic field is applied in the sample plane.

The demagnetization induced by pump pulse, which is also a result of the heating, is mediated by the relatively slow phonon-magnon interaction. As we have shown (see Sec. V A and Fig. 3(b) therein) the demagnetization in the garnet films occurs on the time scale of ~ 500 ps. Therefore, the demagnetization itself cannot contribute to the excitation of the precession. Nevertheless, the demagnetization also occurs in the MA-film. In our experimental geometry the static M_z -component is finite in the whole range of the applied fields (see Fig. 2(a)). Therefore, the demagnetization in MA-film, analogous to that observed in the SA-film, should manifest itself as a slow rotation of the probe polarization. Indeed, as one can see from Fig. 5(b), there is a slow change of the induced probe polarization with a characteristic time of ~ 700 ps. This slow process depends on the sign of the applied field and does not depend on the pump polarization and is stronger in the range of small magnetic field. We argue that this contribution to the signal is the manifestation of the laser-induced demagnetization. The character of the time delay dependence of the z -component of the magnetization is somewhat more intricate than the exponential decay, found in the SA-film (Fig. 3(b)). One of the possible reasons for this is the change of the magnetization value, which follows the function (7), is accompanied by the change of the effective anisotropy field (2). As a result, the change of the M_z can deviate from the exponential behaviour (7).

VI. CONCLUSIONS

In conclusions, we have investigated the interaction between femtosecond laser pulses and thin ferrimagnetic substituted iron garnet films grown on a low-symmetry (210)-oriented GGG substrates. We show experimentally that the impact of a laser pulse results in demagnetization and magnetization precession. The former process occurs on the 500 ps time scale as a result of laser-induced lattice heating and consequent increase of the spin temperature mediated by the phonon-magnon interaction. The magnetization precession, in contrast, is excited on a much shorter time scale. Using pump pulses with various polarizations we demonstrated that the precession is in fact excited via two distinct mechanisms. The first one is the ultrafast inverse Faraday effect, which is a general mechanism found by now in a large number of magnetic dielectrics. Competing with this mechanism, is the change of the growth-induced anisotropy. Lack of the pump polarization dependence, as well as slow relaxation time of this mechanism indicate that there is a thermal change of the magnetic anisotropy, triggered by rapid increase of the lattice temperature. We note, that, in contrast to laser-induced dynamics in magnetic metals, this excitation does not rely on the much slower demagnetization.

The excitation of the magnetization precession in magnetic dielectrics via thermal change of the anisotropy has

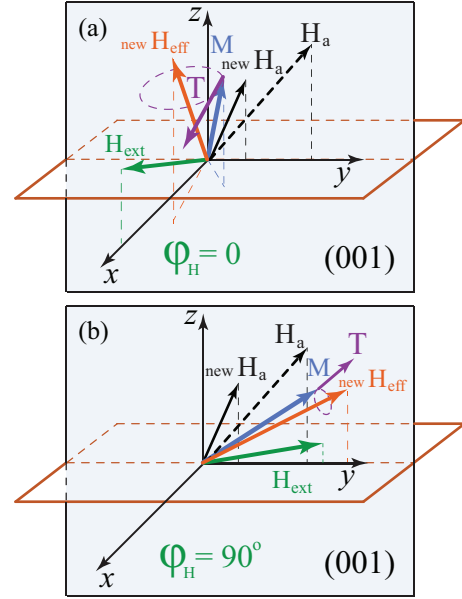


FIG. 8: Graphical illustration of the process of pulse-induced magnetic anisotropy change with the following precessional dynamics for (a) $\phi_H = 0$ and (b) $\phi_H = 90^\circ$. Note, that in the case (b), the effective anisotropy field and the applied field are both lie in the (yz) -plane. As a result, induced change of the anisotropy parameters does not deflect the net effective field away from this plane. This prevents appearance of the T_z -component of the induced torque. By contrast, when the applied field is perpendicular to the yz plane, the same change of the anisotropy modifies both deflection of H_{eff} from the yz plane and its orientation in the plane. As a result, all three components of the induced torque T should be finite.

been previously explored only in the vicinity of the orientation phase transitions, where the anisotropy is strongly temperature dependent. Here we show that even far from the phase transition region the ultrafast heating of the lattice, resulting in the anisotropy change, can effectively excite the magnetization precession. Interestingly, the amplitude of the precession, excited via this mechanism is comparable to that, occurring due to the IFE, which is expected to be important in the studied garnet films. Furthermore, the relative contributions from these mechanisms can be changed by varying the applied magnetic field. As a result, one can gradually control the initial phase of the precession and its sensitivity to the polarization of the exciting laser pulse.

As we demonstrated experimentally and phenomenologically, the parameters of the magnetization precession excited via ultrafast change of the anisotropy are sensitive to the orientation of the applied magnetic field. Most importantly, the initial phase of the precession changes drastically, depending on the angle of the field with the easy plane of the magnetization. This suggests, that the ultrafast change of the magnetic anisotropy should always be considered when one deals with the precession excited by femtosecond laser pulses in a dielectric placed

in an external magnetic field not collinear with the easy magnetization direction.

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